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(54) Method and Apparatus for Rapid Thermal Processing

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(57) 20 Claims

Canada

CCA 3254 (10-89) 41

METHOD AND APPARATUS FOR RAPID THERMAL PROCESSING

This invention generally relates to a new method and apparatus for the fast thermal processing (such as fast pyrolysis, rapid cracking) of carbonaceous materials (feedstock) involving rapid mixing and heat transfer in a novel reactor design. The heat is transferred to the feedstock from hot particulate solids which are injected into the reactor through several inwardly directed, impinging jets. The feedstock is then injected axially into the reactor in the centre of the impinging jets to form a dense turbulent central jet resulting in rapid mixing and heat transfer.

BACKGROUND OF THE INVENTION

Despite falling world oil prices there is still a present need to develop technology to process alternative feedstocks such as coal, lignites, bitumen, heavy oil, oil residues, tar sands, biomass, biomass - derived liquids, and other carbonaceous materials. Biomass is carbonaceous material derived from recently living plants or animals (eg. wood, agricultural residues, food residues, animal manures, municipal solid waste, etc.). Biomass - derived liquids are organic liquids produced from the thermochemical (eg. pyrolysis, liquefaction, gasification, etc.) or biochemical (eg. acid hydrolysis, fermentation, etc.) processing of biomass materials.

The processing of alternative feedstocks to produce heat, chemicals or fuels can be accomplished by a number of thermochemical processes. Such conventional processes are typically equilibrium processes and yield relatively low grade products. For example, combustion is restricted to immediate thermal applications, and gasification normally produces low-energy fuel gas with limited uses. Liquefaction and conventional pyrolysis often produce low yields of valuable liquid or gaseous products. In addition the liquid products which are produced often require considerable secondary upgrading.



Fast non-equilibrium thermal processes such as fast pyrolysis or rapid thermal cracking are one alternative to conventional thermochemical processes. Fast pyrolysis and rapid cracking are methods of imparting a relatively high temperature to carbonaceous material for a very short time and then rapidly reducing the temperature (i.e. quenching) of the products before chemical equilibrium can occur.

Fundamental fast thermal processing research has demonstrated that the production of valuable (non-equilibrium) chemicals, chemical intermediates, light organic liquids and high quality fuel gases, from a variety of carbonaceous feedstocks, can be maximized and selected over lower quality equilibrium products. Rapid heat transfer to the feedstock is a critical parameter in determining product yields and quality, along with other parameters such as reaction temperature and residence time. Fast thermal reactions such as fast pyrolysis and rapid cracking increase the yields of valuable non-equilibrium products at the expense of solid char and heavy organic liquids (i.e. secondary tars). One problem with such systems is to design a practical reactor system which can achieve rapid heating, controlled short residence times and rapid product quenching over a range of operating temperatures for industrial applications.

In order to achieve high heat transfer rates and to maximize reactor selectivity, the mixing time must be a small fraction of the total reaction time, on the order of 15 to 30 milliseconds.

In the past decade considerable effort has gone into the development of new processes for the rapid thermal processing of carbonaceous feedstocks. While all the processes share similar design philosophies (i.e. high heating rates and short residence time) there are significant differences between them. Most important are the differences in the method of heating the feedstock.

In general, the required transfer of heat can be effected by three possible means:

1. Indirect heat transfer through the reactor walls;
2. Director heat transfer with a gaseous or particulate heat carrier;
3. Internal generation through partial combustion of the feedstock.

Internal generation through partial combustion of the feedstock would necessitate the addition of oxygen to the reactor system and consequently such a system will produce undesirable products and reduce yields of preferred products.

While indirect heat transfer may appear to be the simplest method of heating the feedstock, there are several design and operational problems associated with this approach. At high reactor temperatures there can be significant heat flux limitations as well as reactor fouling problems. An alternative approach which minimizes these problems is to supply the required heat directly either with a gaseous or particulate heat carrier.

There are a number of methods known for achieving rapid thermal processing (fast pyrolysis) through direct heat transfer such as a fluidized bed reactor operating at temperatures between 400 and 650°C. (see "Production of Liquids from Biomass by Continuous Fast Pyrolysis" in Bioenergy 84 Volume 3, Biomass Conversion; D.F. Scott and J. Piskorz). The principal drawback for the general use of fluidized beds in extremely rapid, high-temperature thermal processing is the inability to precisely control the residence time. In addition, fluidized bed reactors can not operate at residences times of less than 500 milliseconds.

In addition Occidental Petroleum has patented several reactor systems for the pyrolysis of subbituminous coal.

One of Occidental's designs is disclosed in U.S. Patent 4,102,773. This reactor pyrolyzes carbonaceous material through the use of a particle heat source introduced to the reactor through the use of an overflow 5 weir. The particulate heat source is introduced tangentially to the feedstock. The heat transfer mechanism is primarily a solid to gas to solid mechanism.

Another type of reactor is disclosed in Occidental Petroleum's U.S. Patent 4,108,732. In this 10 reactor the feedstock is introduced axially into the centre of a swirling annular stream consisting of a particulate heat source, without undue mixing of the streams. This type of reactor would suffer from erosion of the reactor walls and there would be little solid to 15 solid heat transfer.

Another previous design of a fast pyrolysis reactor involves the introduction of a gaseous or particulate solid heat carrier through two tangentially opposed jets with axial injection of the carbonaceous 20 feedstock. The mixing and transfer of heat is effected in an entrained flow reactor system. While this design results in satisfactory mixing between the feedstock and heat carrier for a wide range of conditions, several problems were observed. Upon injection, the heat carrier 25 remained concentrated in a thin annular region near the wall and only mixed with the carbonaceous feedstocks at the reactor outlet. This was a direct result of the design of the heat carrier injection system which produced a cyclonic effect within the reactor. In addition, when a 30 particulate heat carrier is used, injection of the solid particles tangentially into the reactor results in severe erosion of the interior walls of the reactor.

A further problem with some prior reactor designs is that the primary mechanism for transferring heat is a 35 solid to gas to solid mechanism. Increased heating rates can be achieved by increasing the amount of solid to solid

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heat transfer. A pyrolysis reactor relying on solid-conductive heat transfer could have over 100 times the throughput of a similarly sized reactor relying on radiative heat transfer as the solid conductive approach transfers energy to the feedstock at a rate over two orders-of-magnitude greater than black body radiation at similar temperatures.

Fast pyrolysis for example appears to proceed initially by the depolymerization, melting and vapourization of a surface layer of the carbonaceous material. If the initial layer is not mechanically removed and is allowed to remain on the feedstock surface, it will absorb a considerable amount of energy as it further pyrolyzes and vapourizes. This protective film adversely affects the ability to transfer heat into the feedstock, and undesirable chemical reactions can occur. If the protective film is mechanically removed by the action of a solid heat carrier for example, greater rates of heating can be achieved. With solid to solid heat transfer, (or solid to liquid if a liquid feedstock is used) the heating rate can thus be increased with less char formation than with solid to gas to solid heat transfer. This type of fast thermal process is an "ablative" mechanism.

Creation of an improved system for fast thermal processing in a manner which is practical for industrial operation is required however. Such a system should incorporate the following features:

- a solid heat carrier
 - a direct solid to feedstock heat transfer
 - rapid and thorough mixing
 - controlled reaction residence time
 - minimal erosion problems
 - minimal deposition or coking on the reactor walls
- 35 An improved system which incorporates the above features and provides rapid heat transfer and thorough mixing

through intimate solid to feedstock contact has been realized.

SUMMARY OF THE INVENTION

Accordingly the invention herein comprises a process and apparatus for achieving efficient rapid thermal processing of carbonaceous feedstocks. In particular an efficient solid heat carrier injection system allows for rapid and effective mixing of the feedstock and solid heat carrier, resulting in intimate contact between the heat carrier and feedstock surfaces and a minimization of equipment erosion problems, through the utilization of non-parallel, impinging jets of particulate heat carrier. Coke deposition is minimized by constricting the feedstock in the reactor core by an envelope of the heat carrier. Specifically the heat carrier is injected through the use of a plurality of radially spaced converging jets directed inwardly and downstream which impinge upon a central jet of feedstock.

In such a system both turbulent shear forces and particle momentum is utilized to achieve rapid mixing. In addition the inward radial momentum of each heat carrier jet is dissipated against the others resulting in reduced equipment erosion.

The mixing of two dispersed particulate phases is effected by turbulent particle dispersion and a large relative velocity difference between the phases. A large relative velocity difference between the two phases causing rapid initial macromixing. Subsequent micromixing is then obtained through turbulent particle dispersion.

Accordingly it was determined that an optimum reactor design would incorporate a converging jet injection system involving two dispersed particulate phases. Carbonaceous feedstock would be injected through the central primary jet and the solid heat carrier, through several secondary converging jets impinging on the central jet of feedstock.

In accordance with the present invention, there is provided a fast thermal processing reactor comprising:

- (a) an elongated conduit reactor;
- (b) first inlet means communicating with the said reactor for axially admitting a primary stream of carbonaceous material;
- 5 (c) second inlet means communicating with the said reactor for admitting a secondary stream of heat supplying particulate material directed radially inwardly and downstream into the said reactor;
- 10 (d) a reacting zone within the said reactor and downstream of the said first and second inlet means where the said secondary stream converges on the said primary stream, and through which the combined streams flow; and
- 15 (e) outlet means in said reactor, downstream of said reacting zone for removing material from the said reactor.

In addition there is provided a process for fast thermal processing of carbonaceous material comprising:

- (a) introducing a primary axial stream of carbonaceous material, into an elongated cavity in a thermal process reactor;
- (b) introducing a secondary stream of particulate heat supplying material, into said elongated cavity, said secondary stream directed inwardly and downstream, said secondary stream converging on said primary stream with the net radial momentum of the secondary stream being virtually zero;
- 25 (c) subjecting the stream of carbonaceous material to the influence of the said heat supplying secondary particulate stream in a reacting zone to cause transformation of the carbonaceous material; and
- 30 (d) removing all materials from said reactor means through outlet means.

Brief Description of the Drawings

35 Details of embodiments of the invention are described by reference to the accompanying drawings:

Figure 1 is a schematic representation of a fast pyrolysis flow system for fast pyrolysis of a solid carbonaceous feedstock employing the reactor of the present invention.

5 Figure 2 is a top plan view of the reactor of one embodiment of the invention.

Figure 3 is a section of the line III-III of Figure 2.

10 Figure 4 is a schematic diagram of a heat carrier feeder used with the reactor of the present invention.

Detailed Description of Preferred Embodiment

In the following description the corresponding elements as shown in each figure of the drawings are given the same reference number.

15 The major components of the fast pyrolysis apparatus are designed to achieve a relatively high temperature within a minimum amount of time as well as having a relatively short residence time at that temperature to effect pyrolysis of a carbonaceous 20 feedstock. Rapid cooling or quenching of the products is required in order to preserve the yields of the valuable equilibrium products.

25 The major components of the fast pyrolysis process incorporating the reactor (ie. thermal mixer) of the invention are illustrated in Figure 1. Rapid mixing and heat transfer are carried out in two vessels. The first vessel, the thermal mixer (1), allows heat to be transferred to a solid particulate carbonaceous feedstock or an atomized liquid carbonaceous feedstock from the hot 30 particulate solid heat carrier (ie. hot silica sand). Mixing and rapid heat transfer occur within 0.10 seconds in this thermal mixer and preferably within 0.015 to 0.030 seconds. The heating rate of the feedstock should be greater than 1000°C per second. The second vessel (2), 35 the quencher allows fast cooling or quenching of the products to a temperature less than 300 C within 0.10

seconds in order to reduce secondary reactions of the initial pyrolysis products exiting from the transport reactor (9). Preferably the pyrolysis products are reduced to below 300°C within 0.030 seconds.

5 The rapid mixing/heat transfer operations of the thermal mixer are separated from the operation of the transport reactor (9). This allows for precise control of the total reaction residence time since the time for heat transfer/mixing is a relatively small fraction at the net
10 residence time. The total residence time in the reactor system (i.e. thermal mixer and transport reactor) is typically in the range of 0.05 to 0.90 seconds.

15 A fluidized bed, for example, can accomplish the heat transfer but there is very limited control of the residence time, and the residence time distribution is broad with a significant portion of the reactants remaining in the reactor for a period longer than the average residence time. A conventional transport or
20 plug-flow reactor, on the other hand, can offer fine control of the residence time but the heat transfer is limited.

25 As shown in figures 2 and 3, the thermal mixer (1) has converging inlets (3) for the solid heat carrier. This system effectively destroys the radial momentum of the heat carrier solids causing severe turbulence. The heat carrier is injected through eight jets which impinge upon a central feedstock jet (4). The ratio of the heat carrier mass flowrate to that of the carbonaceous feedstock depends on the thermal requirements of the
30 system which in turn, depend on the nature of the feedstock and the desired reaction temperature. The preferred range of this ratio is between 5:1 to 50:1. The jet diameters can be selected based upon the desired velocity of the heat carrier and feedstock. The preferred
35 range of velocities is between 5 to 20 metres per second, and the preferred solids loading (i.e. the mass ratio of

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heat carrier solids or feedstock to its carrier gas) is between 10 and 50. Particle size of the carbonaceous feedstock is usually less than 3mm for solids and less than .05 mm for atomized liquids. Typically, the heat carrying solids are inert silica sand with a mean particle size in the range of 50 to 500 microns.

The reactor cross sectional area is approximately equal to that of the jet inlets. An injection angle of 60° was chosen as the preferred angle for the introduction of the heat carrier jets to the central jet.

As shown in figure 1 two types of feeders are used to deliver solids to the reactor system. In the feedstock feeder (6), solid particulate carbonaceous materials passes from a sealed hopper which has a sufficient inventory of feedstock to a funnel system, and are thereby metered onto a rotating table. Two fixed armatures sit near the surface of the rotating table and plow the carbonaceous feedstocks off the outer circumference. The feedstock then falls into a chamber where it is picked up and carried into the transport line by a gaseous carrier. The overall range of the feed rate of the feedstock is controlled by setting the gap between the lower funnel and the table.

The solid heat carrier feeder (7) delivers hot solids to the thermal mixer (1) through a plurality of radially positioned inlets which direct the heat carrier inwardly and downstream into the reactor.

The solid particulate carbonaceous feedstock (or atomized carbonaceous liquids) is then injected axially into the reactor (1) through a water or air cooled tube (18) into the turbulent region where effective mixing and rapid heating to at least 400°C occurs within 0.10 seconds, and preferably within 0.03 seconds.

The fast pyrolysis of carbonaceous feedstock is initiated in the thermal mixer reactor (1) and continues in a transport reactor (9). The transport reactor is a

length of pipe which is housed in electrical oven (10).
The mixture of hot gases and particulate solids passes
from the thermal mixer reactor (1) through the transport
reactor (9) to the quencher (2) and the solids separator
5 (23). The hot gaseous product is rapidly cooled to below
300°C in less than 0.10 seconds and preferably within the
0.03 seconds. The insertion of cylindrical inserts to
reduce the reactor volume, changing the length of the
transport reactor (9) and manipulating the heat
10 carrier/feedstock flow rate, can vary the total residence
time between 30 and 900 milliseconds. Reactor
temperatures can be set in the range of 400° to 1,000°C.

An efficient cyclonic condensor (25) is used to
increase the yield of recovered liquid products. In
15 addition an electrostatic precipitator (24) can be
integrated into a downstream gas line to recover
additional liquid products.

Figure 4 illustrates the design of the heat
carrier feeder. Particulate solids (i.e. silica sand) are
20 contained in a large hopper (7) connected to a stand pipe
(12). At the base of the stand pipe, the solid flow rate
is regulated by a central orifice (13). A relatively long
stand pipe is used to ensure that changes in bed depth
during feeding have a negligible effect on the solids flow
25 rate. To stop the flow of solids a high temperature valve
(14) is used on the transport line underneath the orifice.

From the orifice the solids fall unrestricted
down the stand pipe to a gas/solid mixer (15) where they
are mixed with the transport gas (16). A pressure
30 equalization line (17) connects to the top of the hopper
allowing gas to pass downward through the vent to the
orifice.

The use of a particulate heat carrier in place of
a gaseous heat carrier in a reactor system of this type
35 results in a significant change in the observed jet
structure. Cold model studies using a particulate

feedstock with only a gaseous heat carrier resulted in an observed jet structure that was characteristic of two phase expanding jets. The feedstock jet outlet particles appeared to be evenly distributed over the jet cross section. Downstream the feedstock particles disperse gradually over the entire reactor cross section. The inside expansion angle of the jet was approximately 15°. The jet structure appears very stable.

Upon the addition of a particulate heat carrier to the system however a significant change in the jet structure is observed. The resulting jet structure formed from the use of two dispersed particulate phases, results in a unique solid jet structure formed within the reactor. Shortly downstream of the jet inlets, the feedstock and heat carrier solids combine to form a dense central jet, the diameter of which is considerably smaller than that of the feedstock jet inlet. Thus the jets of the heat carrier particles were efficient in entraining essentially all of the feedstock particles. In addition it appears that the radial momentum of the incoming heat carrier particles was completely dissipated against the central jet of the feedstock since no heat carrier particles traverse through the jet.

Downstream the solids disperse gradually over the reactor cross section.

For the reactor to function properly the radial momentum of each jet must be approximately equal to each other, in order to preserve the balance of radial momentum. While eight jets are shown in figure 3, any number of jets greater than 2 is sufficient as long as the radial momentum is cancelled.

By containing the particulate feedstock in the central core of the reactor during the initial heat up phase of pyrolysis, the feedstock particles are largely prevented from sticking to the reactor wall and forming coke. This is particularly beneficial in dealing with heavy oil or subbituminous coal feedstock.

In addition since the radial momentum of the heat carrier particles are substantially reduced as the jets collide in the centre of the reactor, erosion of a vessel wall will be largely eliminated. The impingement of the feedstock and heat carrier jets also serves to promote intense interaction between the particles, causing rapid ablation of the feedstock particulates and further increases the overall rate of reaction. In addition, strong interaction between the feedstock and heat carrier particles also serves to increase the rate of heat transfer by conduction.

The net effect of the novel reactor design with a solid particulate heat carrier is to increase the amount of solid to solid heat transfer, the speed by which the heat carrier mixes with and heats the feedstock and virtually eliminates erosion of the inside wall of the reactor.

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The embodiments of the invention in which an exclusive property or privilege is claimed are defined as follows:

1. A process for fast thermal processing of carbonaceous material comprising:

(a) introducing a primary axial stream of carbonaceous material, into an elongated cavity in a thermal process reactor;

(b) introducing a secondary stream of particulate heat supplying material, into said elongated cavity, said secondary stream directed inwardly and downstream, said secondary stream converging on said primary stream with the net radial momentum of the secondary stream being virtually zero.

(c) subjecting the stream of carbonaceous material to the influence of the said heat supplying secondary particulate stream in a reacting zone to cause transformation of the carbonaceous material; and

(d) removing all materials from said reactor means through outlet means.

2. A process as claimed in claim 1 where the secondary stream is introduced through a plurality of radially spaced inlets.

3. A process as claimed in claim 2 where:

(a) the heating rate of the carbonaceous material in the reacting zone is greater than 1,000°C per second;

(b) the residence time of the carbonaceous material and the primary products in the reacting zone is between 0.05 seconds and 0.90 seconds;

(c) the temperature of the reacting zone is between 400 and 1000°C; and

(d) the temperature of the products is reduced after removal from the reactor to less than 300°C in less than 0.1 seconds.

4. A process as claimed in claim 2 or 3 where the carbonaceous material is a particulate carbonaceous material.
5. A process as claimed in claim 2 or 3 where the carbonaceous material is an atomized liquid.
6. A process as claimed in claim 3 where the carbonaceous material is ground wood.
7. A process as claimed in claim 3 where the carbonaceous material is coal.
8. A process as claimed in claim 3 where the carbonaceous material is heavy oil.
9. A process as claimed in claim 3 where the carbonaceous material is petroleum derived liquids.
10. A process as claimed in claim 3 where the carbonaceous material is a biomass derived liquid.
11. A thermal process reactor comprising:
 - (a) an elongated conduit reactor;
 - (b) first inlet means communicating with the said reactor for axially admitting a primary stream of carbonaceous material;
 - (c) second inlet means communicating with the said reactor for admitting a secondary stream of heat supplying particulate material directed radially inwardly and downstream into the said reactor;
 - (d) a reacting zone within the said reactor and downstream of the said first and second inlet means where the said secondary stream converges on the said primary stream, and through which the combined streams flow; and
 - (e) outlet means in said reactor, downstream of said reacting zone for removing material from the said reactor.
12. A reactor as claimed in claim 11 where the temperature of the heat supplying particulate material is between 600 and 1100°C.

13. A reactor as claimed in claim 11 or 12 where the reactor reaction temperature is between 400 and 1000°C.
14. A reactor as claimed in claim 11 or 12 where the angle between the first inlet means and the second inlet means is between 30° to 80°.
15. A reactor as claimed in claim 11 where the angle between the first inlet means and the second inlet means is between 50° and 70°.
16. A reactor as claimed in claim 11 wherein the net radial momentum of the second converging stream is about zero.
17. A reactor as claimed in claims 11, 15 or 16 where the second inlet means consists of a plurality inlets, equidistantly spaced.
18. A reactor as claimed in claim 11, 15 or 16 where the second inlet means consists of more than 2 inlets equidistantly spaced.
19. A reactor as claimed in claim 11, 15 or 16 where the second inlet means consists of 3 inlets equidistantly spaced.
20. A reactor as claimed in claim 11, 15 or 16 where the second inlet means consists of 8 inlets equidistantly spaced.

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ABSTRACT

This invention generally relates to a new method and apparatus for the fast thermal processing (such as fast pyrolysis, rapid cracking) of carbonaceous materials 5 (feedstock) involving rapid mixing and heat transfer in a novel reactor design. The heat is transferred to the feedstock from hot particulate solids which are injected into the reactor through several inwardly directed, impinging jets. The feedstock is then injected axially 10 into the reactor in the centre of the impinging jets to form a dense turbulent central jet resulting in rapid mixing and heat transfer.

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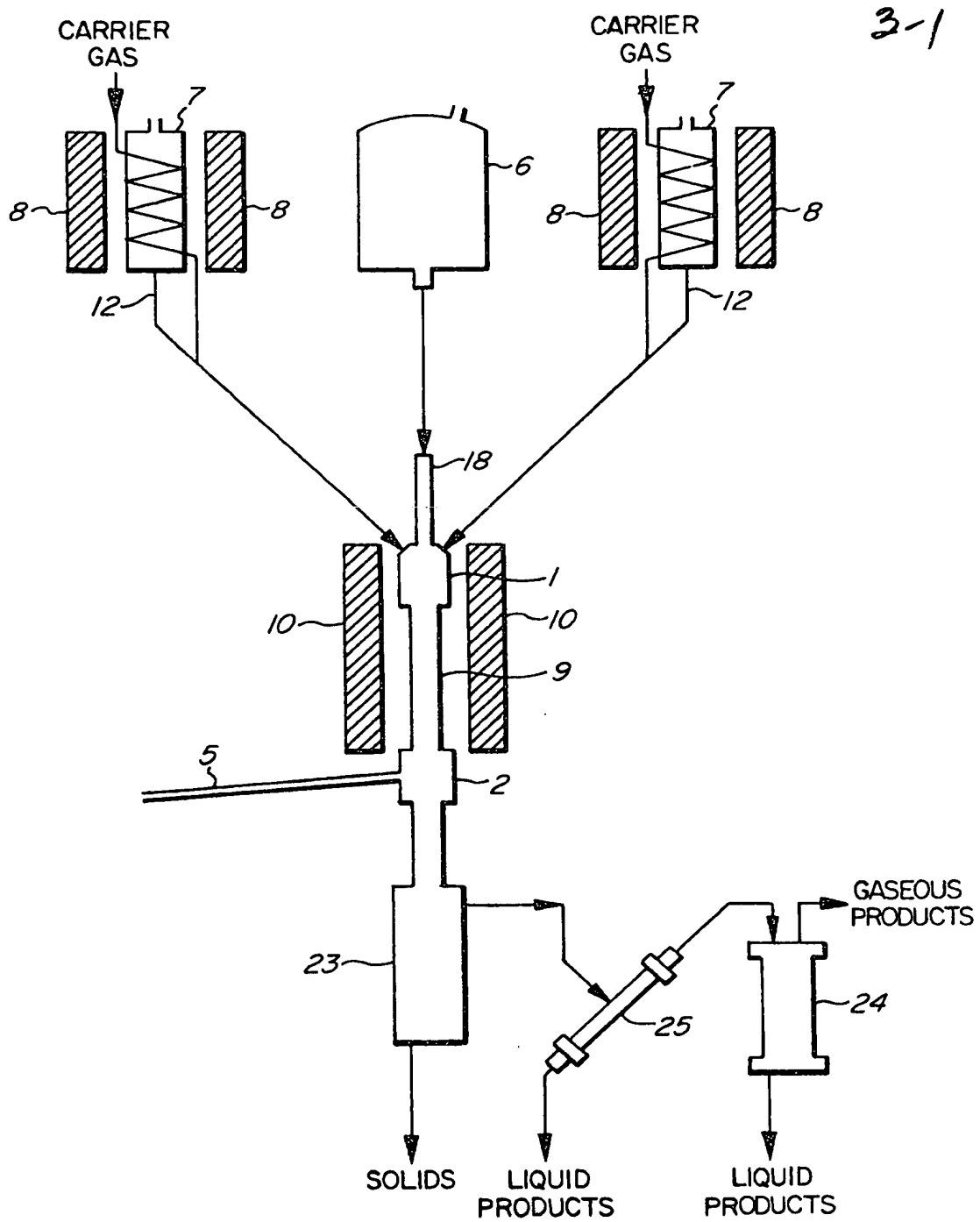


FIG. I

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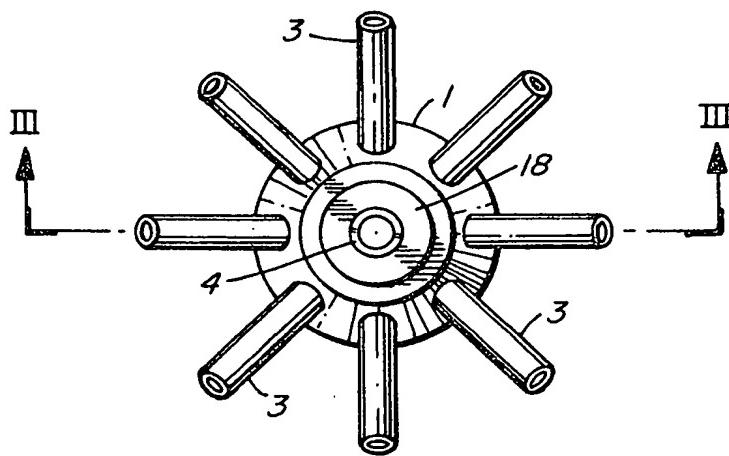


FIG. 2

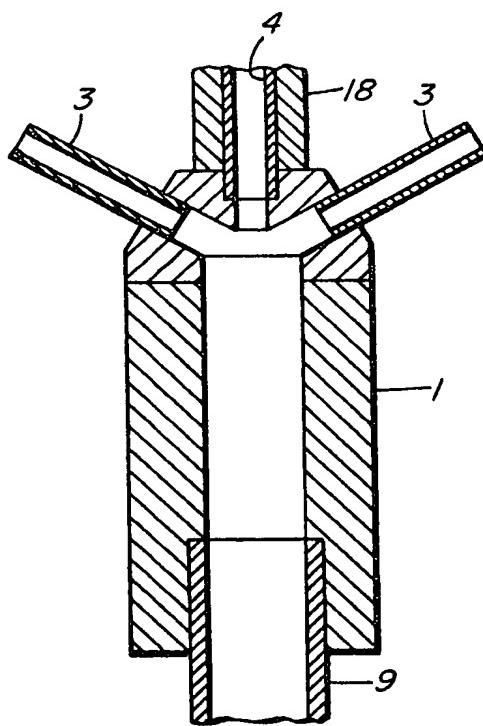


FIG. 3

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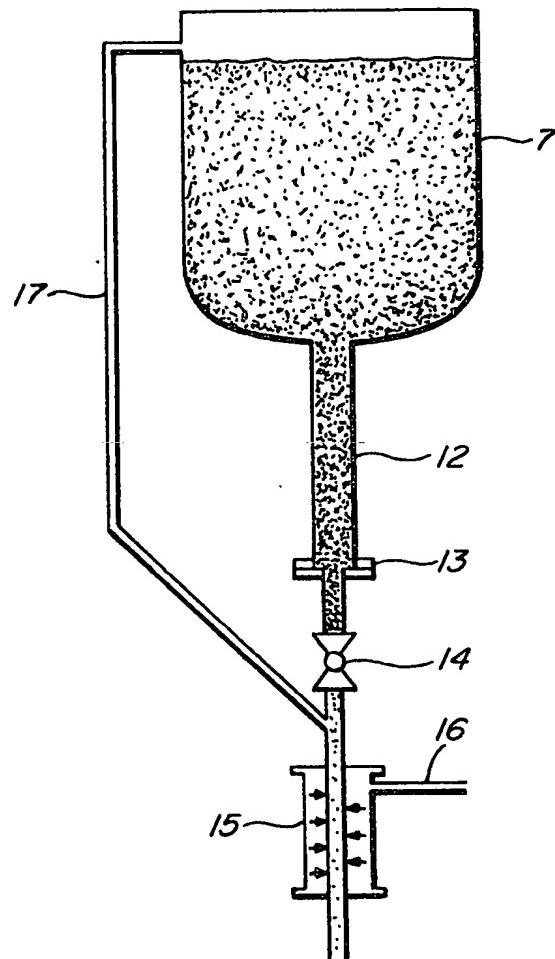


FIG. 4

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